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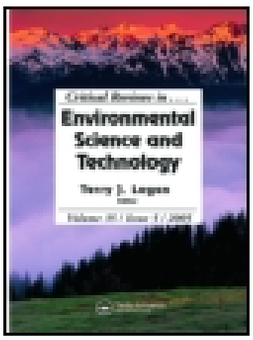

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Recycling nutrients contained in human excreta to agriculture: Pathways, processes, and products

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ABSTRACT

The need for better nutrient management has spurred efforts towards more comprehensive recycling of nutrients contained in human excreta to agriculture. Research in this direction has intensified throughout the past years, continuously unfolding new knowledge and technologies. The present review aspires to provide a systematic synthesis of the field by providing an accessible overview of terminology, recovery pathways and treatment options, and products rendered by treatment. Our synthesis suggests that, rather than focusing on a specific recovery pathway or product and on a limited set of nutrients, there is scope for exploring how to maximize nutrient recovery by combining individual pathways and products and including a broader range of nutrients. To this end, finding ways to more effectively share and consolidate knowledge and information on recovery pathways and products would be beneficial. The present review aims to provide a template that aims to facilitate designing human excreta management for maximum nutrient recovery, and that can serve as foundation for organizing and categorizing information for more effective sharing and consolidation.

KEYWORDS

Phosphorus; nitrogen; potassium; carbon; organic matter; recovery; sewage; wastewater; urine; feces; blackwater; source-separation; fertilizer; soil amendment; resource-oriented sanitation

1. Introduction

Growing concern about future fertilizer availability has re-emphasized the need for better nutrient management, including comprehensive recycling of nutrients contained in human excreta to agriculture (Elser & Bennett, 2009;

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Dawson & Hilton, 2011; McConville et al., 2015). Human excreta have a long history of being used as fertilizer and organic soil amendment but urbanization, the introduction of water closets and sewer networks, and the growing and nowadays widespread use of synthetic fertilizers has contributed to a significant departure from this practice (Rockefeller, 1998; Ferguson, 2014).

In urban areas in industrialized countries, water is used to convey human excreta through extensive sewer networks to municipal sewage treatment plants (STPs). Treatment renders a treated effluent, gaseous emissions, and a solid residual referred to as sewage sludge (in European regulations) or biosolids (in North American regulations). Land application of sewage sludge is a common practice in many countries and allows for partial recycling of nutrients to agriculture. The practice has been heavily debated for a long time, however, due to concerns about contaminants such as pathogens, organic pollutants, and heavy metals in the sludge (Petrik, 1954; Renner, 2000; McBride, 2003; Bengtsson & Tillman, 2004; Singh & Agrawal, 2008; Öberg & Mason-Renton, 2018). As a result, there is a trend towards incineration of a larger portion of the sludge (Kelessidis & Stasinakis, 2012; Kirchmann et al., 2017).

The adequacy and long-term sustainability of conventional urban water and sanitation systems has increasingly been called into question. For low-income countries, the high infrastructure costs are prohibitive for widespread adoption (Larsen et al., 2016). In the context of high-income countries, issues of concern include high energy and water demand, sludge disposal problems, and limited nutrient recycling (Brands, 2014). Some scholars hold on to the idea of municipal sewers and call for more comprehensive resource recovery at municipal STPs (Peccia & Westerhoff, 2015; Verstraete et al., 2016; Puyol et al., 2017). Other scholars hold that source separation and control provide greater opportunities for resource recovery, as it minimizes dilution and contamination of human excreta (Larsen & Gujer, 1997; Otterpohl et al., 1997; Wilsenach et al., 2003; Larsen et al., 2009). Approaches based on source separation and control are commonly referred to as new, ecological, resource-oriented, source-separating, or decentralized sanitation or wastewater management.

Overall, significant research and development has taken place in recent decades to enable more comprehensive recovery of nutrients contained in human excreta. New knowledge and technologies are continuously unfolding, as evidenced by the number and scope of recent reviews published in the scientific literature, see Table 1. These reviews provide detailed insights into certain aspects of nutrient recovery. It is, however, challenging to identify broad patterns and opportunities in the field as a whole, when technical details or certain technologies are studied in isolation.

Table 1. Examples of previous reviews on the recovery of nutrients found in human excreta and streams containing human excreta.

Technology	Reference(s)
Phosphorus recovery	Balmér (2004); Cornel and Schaum (2009); Petzet and Cornel (2011); Rittmann, Mayer, Westerhoff, and Edwards (2011); Sartorius et al. (2012); Desmidt et al. (2015); Egle et al. (2015); Karunanithi et al. (2015); Cieślik and Konieczka (2017); Melia et al. (2017)
Struvite crystallisation	Doyle and Parsons (2002); Le Corre, Valsami-Jones, Hobbs, and Parsons, (2009); Rahman et al. (2014); Kumar and Pal (2015); Darwish, Aris, Puteh, Abideen, and Othman (2016)
Membrane separation	Lutchmiah, Verliefde, Roest, Rietveld, and Cornelissen (2014); Xie, Shon, Gray, and Elimelech (2016); Ansari, Hai, Price, Drewes, and Nghiem (2017)
Sorption	Wang and Peng (2010); Loganathan, Vigneswaran, Kandasamy, and Bolan (2014)
Ecological sanitation	Winker, Vinnerås, Muskolus, Arnold, and Clemens (2009); Haq and Cambridge (2012); Roy (2017)
Biological systems	Nancharaiah et al. (2016); Puyol et al. (2017)
Phototrophic biomass growth	Hülsen, Batstone, and Keller (2014); Sukačová and Červený (2017); Abinandan, Subashchandrabose, Venkateswarlu, and Megharaj, (2018); Santos and Pires (2018)
Bioelectrochemical systems (BES)	Kelly and He (2014); Rodríguez Arredondo et al. (2015); Nancharaiah et al., (2016)
BES applied to urine	Ledezma, Kuntke, Buisman, Keller, and Freguia (2015)
Nutrient recovery from urine	Maurer et al. (2006); Pronk and Koné (2009)
Nutrient recovery from digestate	Monfet, Aubry, and Ramirez (2017); Vaneeckhaute et al. (2017
Nutrient recovery from waste water	Batstone, Hülsen, Mehta, and Keller (2015); Mehta et al. (2015)
Nutrient recovery from sewage sludge incineration ash (SSA)	Donatello and Cheeseman (2013)

These reviews have generally focused on a specific nutrient (notably phosphorus), recovery technology (e.g. membrane separation), input stream (e.g. urine), or a combination thereof. Note that this list is not exhaustive.

The present review aims to provide a rigorously informed and systematic synthesis of available and proposed recovery pathways designed to facilitate recycling of nutrients contained in human excreta to agriculture, covering treatment processes as well as products rendered by treatment. Our aspiration is to present the material in a way that is accessible across diverse yet relevant fields of expertise. The focus is on highlighting broad patterns and opportunities in the field as a whole, and to point to literature that specifically describes certain selected aspects, technologies, or products in more detail.

Most importantly, we hope to facilitate communication and cross-fertilization not only among the various engineering groups that work on the recovery of nutrients found in human excreta, but also between these groups and research communities active in the fields of soil sciences and food and farming systems, as well as other related fields such as industrial ecology, urban metabolism, circular economy, and environmental systems analysis.

2. Human excreta

As our intention is to write for a diverse audience, we start by providing a short description of human excreta and how they may get mixed with other streams prior to treatment. To clarify what it is that treatment aims at

recovering or removing, we also describe factors that impact the composition of different streams that consist of, or contain human excreta, and can form the starting point for the recovery of resources contained in human excreta.

2.1. Carbon and nutrient content of human urine and faeces

Human urine consists of more than 90% water (H_2O) by weight, the remainder being inorganic salts and organic compounds (Rose et al., 2015). The dried solids contain about 13% carbon (C), 14–18% nitrogen (N), 3.7% phosphorus (P), and 3.7% potassium (K) (Rose et al., 2015). Urea (CH_4N_2O) is the dominant solute in fresh urine, making up over 50% of the organic compounds (Rose et al., 2015). About 85% of N is fixed in urea and about 5% as total ammonia (NH_3 and NH_4^+) (Udert, Larsen, & Gujer, 2003, Udert, Larsen, & Gujer, 2006). Shortly after urination, the nonvolatile urea is broken down into bicarbonate (HCO_3^-) and carbonate (CO_3^{2-}) as well as nonvolatile ammonium (NH_4^+) and volatile ammonia (NH_3) (Udert, Larsen, & Gujer, 2003). Urea hydrolysis is a spontaneous process because the bacteria that produce the urea hydrolyzing enzyme urease are ubiquitous (Udert, Larsen, Biebow, & Gujer, 2003). After urea hydrolysis, about 90% of total N in urine is present as ammonia or ammonium (Udert et al., 2006). Urea hydrolysis implies the potential for ammonia volatilization during collection, storage, transport and application of urine, especially because the pH can increase up to 9 during the process, shifting the equilibrium from nonvolatile ammonium to volatile ammonia (Hellström et al., 1999; Chang et al., 2015). Human feces consist of about 75% H_2O by weight and 25% solid material, mainly organic matter (Rose et al., 2015). C is a major constituent of the dried solids as approximately half of organic matter generally is C (Vassilev et al., 2010) and this is also true for feces (Rose et al., 2015). N, P, and K make up 5–7%, 3–5.4%, and 1–2.5% of the dried solids respectively (Rose et al., 2015). Both urine and feces also contain a range of micronutrients such as magnesium (Mg) and selenium (Se). The amount of excreted nutrients depends on dietary intake, while the digestibility of the diet determines the partitioning of nutrients between urine (digested) and feces (undigested) (Jonsson et al., 2004). Generally, urine contains the majority of N and about half of P and K contained in human excreta, while feces are rich in P and K and contain the majority of C (Heinonen-Tanski & Van Wijk-Sijbesma, 2005).

2.2. Contaminants of concern in human urine and faeces

Human excreta commonly contain pathogens. Faeces always contain high numbers of enteric bacteria (e.g. *Campylobacter*, *Salmonella*) and may also contain high numbers of viruses (e.g. Norovirus, Rotavirus), protozoa

(e.g. *Cryptosporidium*, *Giardia*), and parasitic worm eggs (e.g. *Ascaris*) (Heinonen-Tanski & Van Wijk-Sijbesma, 2005). Fresh urine, especially from healthy persons, contains few pathogens (Heinonen-Tanski & Van Wijk-Sijbesma, 2005; Udert et al., 2006). Faecal cross-contamination of urine during and after excretion, however, can increase the number of pathogens in urine (Jönsson et al., 1997; Schönning et al., 2002). Human excreta can also contain heavy metals and organic pollutants, notably pharmaceutically active substances such as pharmaceuticals, pharmaceutical residues, and (synthetic) hormones. Heavy metal concentrations in urine are generally very low in relation to the nutrients; feces constitute a much higher heavy metal load compared to urine (Jönsson et al., 1997; Tervahauta et al., 2014). Some of the organic pollutants are mainly excreted with urine, while others are excreted mostly with feces (Lienert et al., 2007).

2.3. Mixing of human excreta with other streams

Collection of human excreta often involves mixing with other streams (see Figure 1). Separate collection of urine, depending on the type of toilet or urinal, may involve mixing with flush water and the respective stream is commonly referred to as source-separated urine or yellowwater. Separate collection of human feces, depending on the type of toilet, may involve mixing with urine, flush water, anal cleansing water, toilet paper, and additives such as ash, lime, or dried soil. The respective stream is commonly

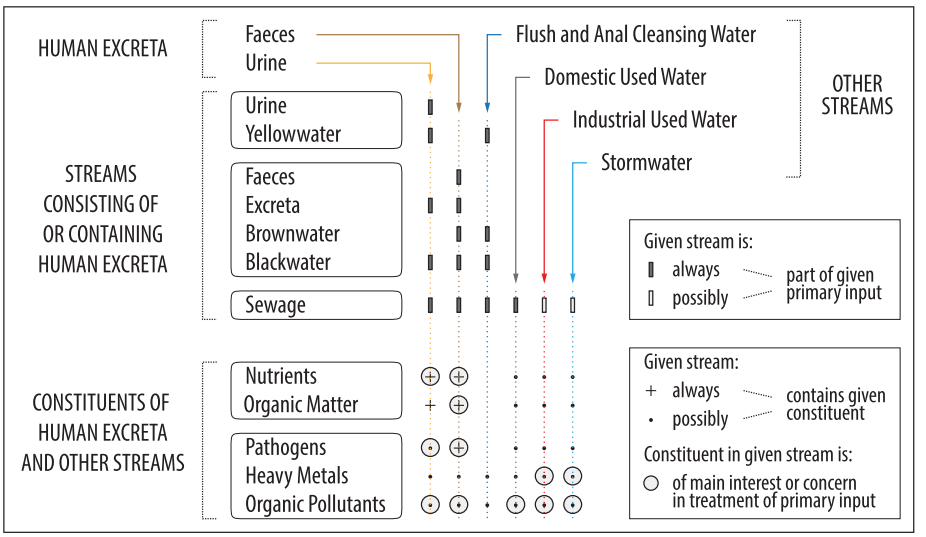

Figure 1. Overview of: (1) how human excreta and other streams are combined into a range of primary inputs that form the starting point for recovery pathways reported in peer-reviewed studies dealing with recovery of resources from human excreta; and (2) constituents of interest or concern in human excreta and other streams. Note that used diapers are not considered in the present review, even though they also represent a stream containing human excreta.

referred to as source-separated feces, brownwater, excreta, or blackwater. In conventional urban water and sanitation infrastructures, human excreta generally become mixed with flush water, anal cleansing water, toilet paper, domestic used water, industrial used water, and possibly even stormwater. The stream resulting from this form of collecting human excreta is commonly referred to as domestic or municipal sewage or wastewater.

2.4. Sources of carbon, nutrients, and contaminants in mixed streams

Human excreta normally are the major contributor of nutrients and organic matter in any of the streams containing human excreta, although the amount of nutrients and organic matter will increase if organic kitchen refuse is collected through the same collection system or added to treatment as supplemental feedstock (Kujawa-Roeleveld et al., 2006; Friedler et al., 2013). Flushwater can add heavy metals and organic pollutants originating from the water supply system, as for example copper (Cu) and lead (Pb) can be released from metal pipes (Renner, 2008; Schock et al., 2008) or organic compounds from polymeric pipes (Zhang & Liu, 2014). Contamination levels are further increased following mixing with used water from households, hospitals, industry, and the commercial sector, and with stormwater where it is also discharged to the same sewer. Pathogens mainly originate from human excreta (Dumontet et al., 2001), but can also originate from meat preparation in domestic kitchens (e.g. *Salmonella* and *Campylobacter* during the preparation of chicken) (Cogan et al., 1999) or commercial processing of animal products (e.g. in tanneries, meat markets, abattoirs) (Dumontet et al., 2001). Organic pollutants include substances such as pharmaceutically active coumpounds and hormones, personal care products, and detergents (Kümmerer, 2013). Pharmaceutically active compounds and hormones mainly originate from human excreta whereas other organic pollutants mainly originate from other sources. Heavy metals originate from different sources (Sörme & Lagerkvist, 2002). Compared with other sources of heavy metals, those contained in human excreta typically account for less than a tenth of total load in sewage (Tervahauta et al., 2014).

3. Recovery pathways

Efforts to recover resources from human excreta or streams containing human excreta have typically targeted water, energy, carbon, nutrients, metals, or a combination of these resources.

Here, we compile and describe recovery pathways that facilitate nutrient recovery. Given the iterative nature of the literature search and analysis, information about what we found is presented along with how we found it.

3.1. Conceptual model and terminology

A simple input-output model (Figure 2) guided our literature analysis and is useful to explain central terms. We define as primary input any stream that contains human urine and/or feces and that forms the starting point for resource recovery. Treatment aims to facilitate recovery and recycling of resources found in the primary input. Where treatment comprises more than one treatment process, the output from one process can become the input to another. Products are defined as outputs that do not become the input to another treatment process. We refer to a specific sequence of treatment processes as a treatment train. A treatment train either transforms a primary input into one single product, or into a number of different products. The combination of a certain primary input, a certain treatment train, and a certain product we refer to as a recovery pathway. Where multiple products are obtained from the same primary input and treatment train, each product comprises a separate recovery pathway.

3.2. Recovery pathways facilitating nutrient recovery

We searched the scientific literature for documents describing recovery pathways specifically targeted towards nutrient recovery through the search strategies described in Supporting Information 1. In doing so, we iteratively identified and developed categories for treatment processes and products rendered by treatment. Recovery pathways were arranged into clusters of pathways that start from similar primary inputs, feature similar treatment processes, and/or render similar products. These clusters as well as variations within clusters and a list of documents constituting each cluster are detailed in Supporting Information 1. A simplified representation of common recovery pathways is shown in Figure 3 and explained in the remainder of this section.

Treatment trains starting from urine or yellowwater represent two broad strategies. The first strategy applies treatment processes that aim at

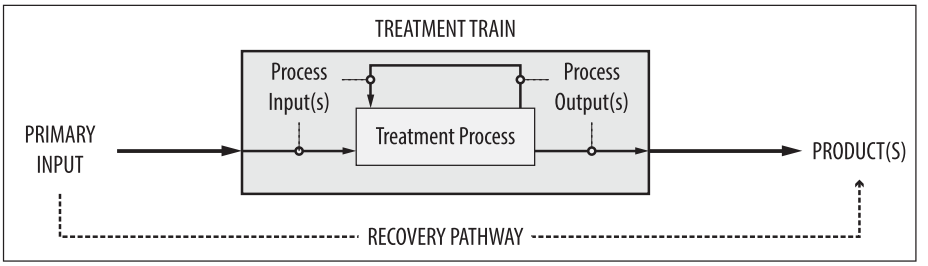

Figure 2. Simple input-output model that guided our analysis of peer-reviewed scientific studies dealing with resource recovery from human excreta and streams containing human excreta.

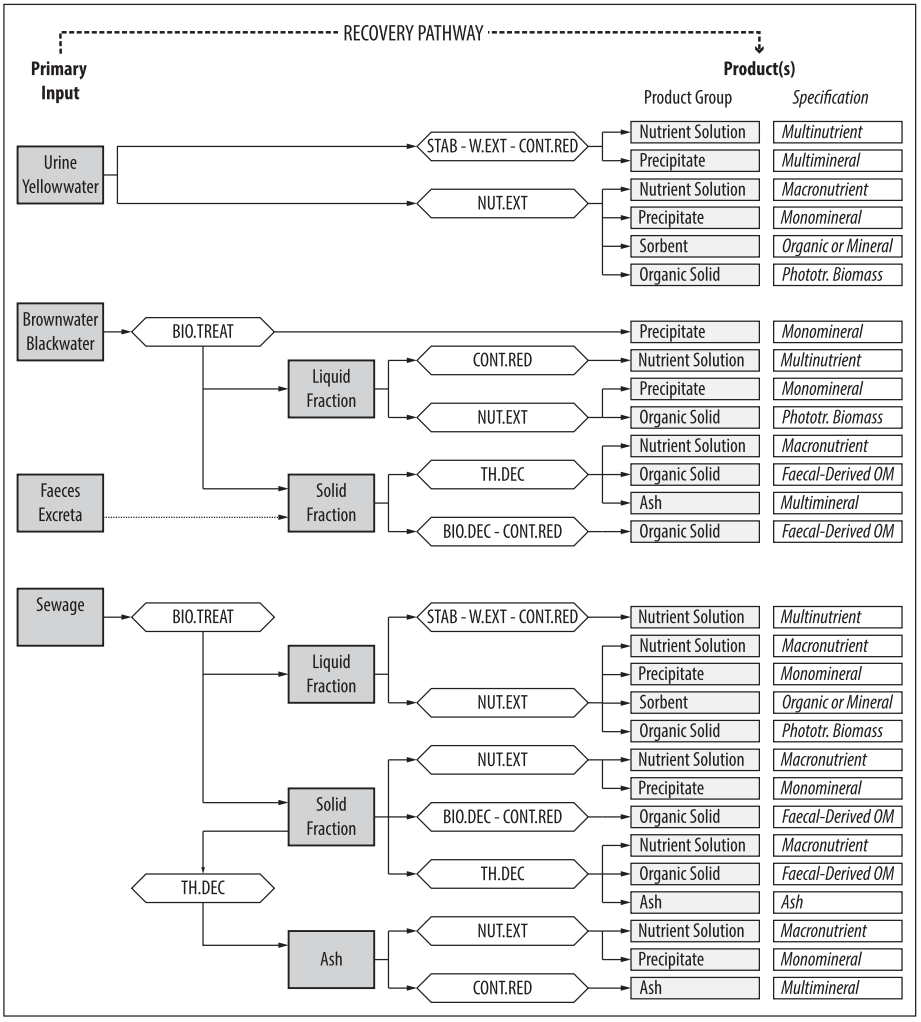

Figure 3. Simplified representation of recovery pathways targeted towards nutrient recovery from human excreta and streams containing human excreta, as reported in the peer-reviewed scientific studies included in the present review. Abbreviations: BIO.TREAT = biological treatment; BIO.DEC = biological decomposition; TH.DEC = thermal decomposition; STAB = stabilization; W.EXT = water extraction; CONT.RED = contaminant reduction; NUT.EXT = nutrient extraction. A more comprehensive representation can be found in Figure S1.1 in Supporting Information 1. For further explanation of product groups and specifications the reader is referred to chapter 5.

prevention of ammonia volatilization, separation of water from nutrients, and/or contaminant reduction (through separation of contaminants from nutrients and/or the pathogen inactivation and/or organic pollutant degradation). The second strategy is characterized by selective nutrient extraction. Treatment processes applied to this end often also imply volume reduction (through separation of nutrients from water) and contaminant reduction (through separation of nutrients from contaminants) (Maurer et al., 2006).

Treatment trains starting from brownwater or blackwater commonly begin with (anaerobic) biological treatment followed by liquid-solid separation. Biological treatment can be designed such as to enable simultaneous nutrient extraction, for instance through precipitation or granulation. The liquid fraction can be the input to processes aimed at depollution (notably pathogen inactivation) or nutrient extraction. The solid fraction, or the dryer primary inputs feces and excreta, can be the input to depollution (notably pathogen inactivation), or to biological decomposition (possibly enhanced by additional pathogen inactivation) or thermal decomposition of organic matter.

Treatment trains starting from sewage commonly involve liquid-solid separation, usually preceded by or as a part of (aerobic) biological treatment. The liquid fraction or effluent can be the input to processes aimed at contaminant reduction or nutrient extraction. The solid fraction (sewage sludge) can be the input to processes aimed at contaminant reduction (pathogen inactivation), nutrient extraction, and/or biological or thermal decomposition of organic matter. Ash, the inorganic residual rendered by some thermal decomposition processes, can be the input to processes aimed at nutrient extraction or contaminant reduction (heavy metal removal).

Note that there are certain thermal decomposition processes that have generally been targeted towards recovering carbon in the form of energy carriers rather than nutrient recovery. Processes of this kind include hydrothermal carbonization (HTC) (Danso-Boateng et al., 2013; Danso-Boateng, Holdich, Martin, Shama, & Wheatley, 2015; Danso-Boateng, Shama, Wheatley, Martin, & Holdich, 2015), hydrothermal gasification (HTG) (Afif, Azadi, & Farnood, 2011; He, Chen, Giannis, Yang, & Wang, 2014), and gasification (Rong et al., 2015) with feces or sewage sludge as feedstock. We have included these processes because they can facilitate nutrient recovery, in principle.

3.3. Developing the option space for nutrient recovery

Mapping recovery pathways quickly becomes subject to redundancy, even in a simplified representation like Figure 3, because different primary inputs can be subjected to similar treatment trains, and different treatment trains can feature similar treatment processes and render similar products. Our aspiration was to produce a map of primary inputs, treatment processes, products rendered by treatment, and their relationships. This map we refer to as the 'option space' for nutrient recovery. To avoid redundancy, we identified four broad categories of similar process inputs and outputs (including primary inputs and products). We mapped these four categories of process inputs and outputs, indicating how treatment processes can convert an input belonging to one category to an output belonging to the same or a different category. This resulted in a refined input-output model

that forms the backbone of the option space for nutrient recovery. The step-wise process leading to the option space is illustrated in Figure S2.1 in Supporting Information 2. The actual option space is shown in Figure 4. Note that the option space allows for the output from one process to become the input to a following process. The option space is a generic representation that can map any recovery pathway that builds on the primary inputs, treatment processes, and products featured as building blocks of the option space. Treatment processes are described in more detail in Section 4, products in Section 5.

4. Treatment processes

A brief description of each treatment process featured in the option space, a simple input-output diagram, and details regarding the fate of constituents (nutrients, organic matter, pathogens, organic pollutants, and heavy metals) during treatment are provided in Supporting Information 3. Here, we summarize treatment processes and the fate of constituents during treatment.

4.1. Decomposition of organic matter

Decomposition refers to the breakdown of organic matter into smaller and more stable molecules, which can be achieved via biotic (biological) or abiotic (mechanical, thermal, chemical, or thermo-chemical) processes (Atay & Akbal, 2016). As for biotic processes, we here distinguish biological treatment and biological decomposition. Biological treatment refers to processes where the solid retention time (SRT) is larger than the hydraulic retention time (HRT), that is, the solid fraction of the input stays in the system longer than the liquid fraction. Biological decomposition refers to processes where solid and liquid fraction stay in the system for the same period of time (SRT equals HRT).

4.1.1. Biological treatment

The activated sludge process, invented roughly a century ago, is still at the core of many contemporary municipal STPs and was originally designed to remove organic matter from municipal sewage or industrial wastewaters (Orhon, 2015). Over the years, biological N and biological or chemical P removal processes have been incorporated into overall process design to meet ever stricter effluent standards aimed at minimizing the release of N and P to the aquatic environment (Cooper, 2001). In tropical climates, anaerobic treatment of sewage is a frequently applied alternative to the activated sludge process (Seghezzo et al., 1998). The upflow anaerobic sludge blanket (UASB) reactor, developed in the 1980s (Lettinga et al., 1980), is

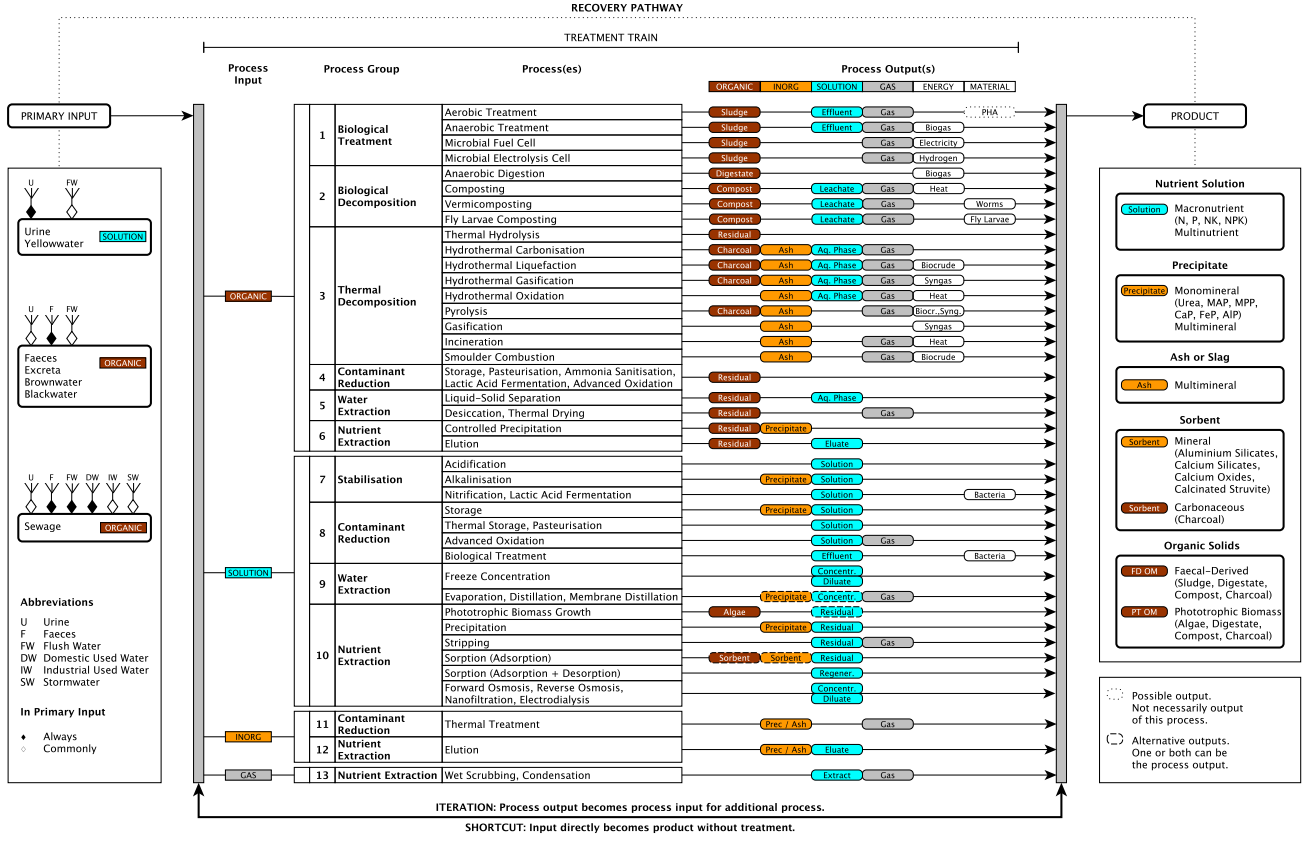

Figure 4. Option space for nutrient recovery. Note that the option space presented here can easily be extended to accommodate additional primary inputs, products, and treatment processes, in order to also map novel recovery pathways.

the most applied anaerobic system for treatment of sewage and industrial wastewaters. Also blackwater is succesfully treated applying UASB technology (de Graaff et al., 2010; Hernández Leal et al., 2017). Bioelectrochemical systems such as microbial fuel cells (MFCs) and microbial electrolysis cells (MECs) represent an emerging technology for treating wastewater under anaerobic conditions and can be applied to urine, low-strength wastewater such as municipal sewage, as well as high-strength industrial wastewaters (Gude, 2016). While initially designed for efficient wastewater treatment, nutrient recovery has become an integral part of process design (Kelly & He, 2014; Nancharaiah et al., 2016; Goglio et al., 2019). More recently, treatment configurations that enable the formation of polyhydroxyalkanoates (PHA) or other precursors for high-value products have received increased attention (Morgan-Sagastume et al., 2014; Modin et al., 2016; Pittmann & Steinmetz, 2017; Puyol et al., 2017). The decomposition of organic matter releases nutrients from the organic matter to the liquid phase, adding to those nutrients already present in their dissolved form. Process designs based on the activated sludge process can achieve much lower concentrations of N and P in the effluent compared to anaerobic treatment (Seghezzo et al., 1998). Retaining nutrients in the effluent is beneficial if the effluent is used for fertigation in agriculture. Treatment under anaerobic conditions avoids consuming energy for aeration. Instead, energy is recovered in the form of biogas (in anaerobic treatment such as UASB) (Seghezzo et al., 1998), electricity (in microbial fuel cells) or biofuels such as ethanol, methane, or hydrogen (in microbial electrolysis cells). Pathogens tend to only partly accumulate in the sludge (Wen et al., 2009; Li et al., 2015; Huang et al., 2018). In aerobic treatment, heavy metals partition fairly equally between effluent and sludge (Karvelas et al., 2003). In anaerobic treatment, heavy metal precipitation to the sludge is generally higher due to sulfide precipitation (Cowling et al., 1992; De la Varga et al., 2013). The partitioning behavior of organic pollutants depends on the compound, with a tendency towards sorbing to the sludge (Katsoyiannis & Samara, 2005). In addition, both aerobic and anaerobic treatment schemes have the potential to inactivate some pathogens and degrade some organic pollutants (Butkovskyi, Rijnaarts, Zeeman, & Hernandez Leal, 2016). Bioelectrochemical systems in particular have been shown to have the potential for high removal of recalcitrant pollutants (Huang et al., 2011).

4.1.2. Biological decomposition

Anaerobic digestion enables recovery of energy in the form of biogas and nutrients in the form of digestate. Composting renders a soil amendment while vermicomposting and fly larvae composting render a soil amendment as well as worms or fly larvae for potential use as animal feed. These

processes are commonly applied to more concentrated streams such as feces or excreta, or fecal, blackwater, or sewage sludge.

As in biological treatment, nutrients are released from organic matter upon its decomposition. If biological treatment takes place in an open system, as often the case for composting and usually the case for vermicomposting and fly larvae composting, volatile forms of N can be lost to the atmosphere, and soluble nutrients to a liquid leachate (Ulén, 1997; Jonsson et al., 2004; Lalander et al., 2015; Nigussie et al., 2016). Biological decomposition can inactivate some pathogens (especially when temperatures above 60 °C are achieved) (Gajurel et al., 2007; Lalander et al., 2013), decompose some organic pollutants (Butkovskyi, Ni, Hernandez Leal, Rijnaarts, & Zeeman, 2016), and influence heavy metal speciation (He et al., 2016).

4.1.3. Thermal decomposition

Thermal decomposition processes can be geared towards facilitating further treatment or safe disposal of an organic feedstock, but can also be designed to facilitate the recovery of resources such as energy, carbon, nutrients, and/or metals. Thermal hydrolysis and advanced oxidation processes (AOPs) such as ozonation aim to make wet organic matter, usually sewage sludge, more biodegradable and are commonly applied as pretreatment to anaerobic digestion (Barber, 2016). Hydrothermal carbonization (HTC) (Danso-Boateng et al., 2013; Danso-Boateng, Holdich, et al., 2015; Danso-Boateng, Shama, et al., 2015), liquefaction (HTL) (Aida et al., 2016; Lu et al., 2017), and gasification (HTG) (Afif et al., 2011; He et al., 2014) aim to convert wet organic matter into charcoal, biocrude, or syngas, respectively. Hydrothermal oxidation (HTO) processes such as low pressure wet oxidation (LOPROX) (Blöcher et al., 2012) and supercritical water oxidation (SCWO) (Stendahl & Jäfverström, 2004) aim at complete destruction and conversion of wet organic matter to carbon dioxide. Pyrolysis (Bridle & Pritchard, 2004; Shepherd et al., 2016; Bai et al., 2017) aims to convert dry organic matter into charcoal and/or biocrude. Gasification (Rong et al., 2015) aims to convert dry organic matter into syngas. Incineration (Li et al., 2017) usually involves complete decomposition of organic matter by means of oxidation to carbon dioxide. Smoulder combustion (Yermán et al., 2015, 2017; Fabris et al., 2017) also aims at complete decomposition of organic matter but can be designed to yield pyrolysis products such as biocrude. Pyrolysis and HTL have been investigated with recovery of both nutrients and energy in mind, while the other processes have typically been targeted primarily towards recovering carbon in the form of charcoal or energy carriers.

Hydrothermal processes (i.e. HTC, HTL, HTG, and HTO) generally yield an inorganic residual in addition to the target product(s). This residual generally consists of a liquid fraction and a solid fraction, which can be

separated from one another by means of liquid-solid separation. Nutrients are partitioned to the carbonaceous target product as well as the inorganic (liquid or solid) residual (Stendahl & Jäfverström, 2004; Blöcher et al., 2012; Kruse et al., 2016; Yao et al., 2016; Lu et al., 2017). Monovalent ions (e.g. NH_4^+ and K^+) tend to partition to the liquid fraction of the inorganic residual, multivalent ions (e.g. PO_4^{3-} and most metal ions) to the solid fraction (Toufiq Reza et al., 2016). Pyrolysis, gasification, and incineration are subject to N volatilization, while P and K as well as most metals are retained in the char or ashes, respectively (Bridle & Pritchard, 2004; Hossain et al., 2011; Gorazda et al., 2018). Pathogens are generally fully inactivated while organic pollutants are partly or fully decomposed depending on process conditions and type of compound (Libra et al., 2011).

4.2. Stabilisation processes

Stabilisation of urine and other liquid streams such as treated effluent is specifically directed to prevent volatilization of ammonia as this can help avoid N losses and negative impacts associated with released ammonia gas such as odor nuisance (Hellström et al., 1999) and acidification of soils and water bodies (Hunter et al., 2011). Stabilisation of urea-rich solutions (e.g. fresh urine) aims at preventing urea hydrolysis and hence preserving N in the form of nonvolatile urea. Stabilisation of ammonia-rich solutions (e.g. urine after urea hydrolysis) aims at converting volatile ammonia to ammonium and other nonvolatile forms of N. While stabilization can be applied as standalone process, it is typically applied in combination with other processes such as storage (e.g. Hellström et al., 1999), or as pretreatment to other processes, notably evaporation (e.g. Senecal & Vinnerås, 2017), distillation (e.g. Fumasoli et al., 2016), membrane distillation (e.g. Tun et al., 2016) and phototrophic biomass growth (e.g. Coppens et al., 2016). Processes geared towards stabilization of liquid streams that have gained most traction include chemical processes such as acidification (e.g. Hellström et al., 1999) and alkalinisation (e.g. Randall et al., 2016) as well as biological processes such as partial nitrification (e.g. Sun et al., 2012) and lactic acid fermentation (e.g. Andreev et al., 2017). Stabilisation processes generally have some potential to inactivate some pathogens (Hellström et al., 1999; Bischel et al., 2015; Randall et al., 2016). Biological processes in addition also have the potential to degrade some organic pollutants (Fumasoli et al., 2016; Andreev et al., 2017).

4.3. Separation processes

The main purpose of separation processes is to separate various constituents in the process input from one another. Several treatment processes

have been investigated for liquid streams such as urine or treated effluent, aiming at separating water and/or contaminants from nutrients, or nutrients from water and/or contaminants.

4.3.1. Freeze concentration

Processes geared towards water extraction from liquid streams include freeze concentration, that is, the concentration of a solution through freezing and melting. Freeze concentration has the potential to retain most nutrients in the concentrate (Lind et al., 2000; Gulyas et al., 2004).

4.3.2. Vaporisation and membrane separation

Processes based on vaporization and/or membrane separation include passive evaporation (e.g. Pahore et al., 2010; Bethune et al., 2014, Bethune, Chu, & Ryan, 2016; Dutta & Vinnerås, 2016), thermal (e.g. Ek et al., 2006; Senecal & Vinnerås, 2017) or solar thermal evaporation (e.g. Antonini et al., 2012), high temperature (Jiang et al., 2017) or low pressure distillation (e.g. Udert & Wächter, 2012; Fumasoli et al., 2016), membrane distillation (MD) (e.g. Tun et al., 2016), forward osmosis (FO) (e.g. Ek et al., 2006; Zhang et al., 2014; Liu et al., 2016), reverse osmosis (RO) (e.g. Ek et al., 2006), and nanofiltration (NF) (e.g. Pronk, Palmquist, Biebow, & Boller, 2006; Lazarova & Spendlingwimmer, 2008). These processes can enable the separation of water from nutrients in liquids as well as liquid-solid separation in slurries that also contain particulate organic matter or minerals. In thermal drying of sewage sludge (e.g. Horttanainen et al., 2017), for example, water is extracted from a slurry through evaporation. Separation of the liquid and solid fractions can also be achieved through membrane separation processes such as FO, RO and NF, as well as a number of other processes such as centrifugation or sedimentation.

N losses can occur due to ammonia volatilization during vaporization (Bethune et al., 2016; Dutta & Vinnerås, 2016; Tun et al., 2016; Jiang et al., 2017), and due to low rejection of urea, ammonia, nitrite and nitrate in membrane separation (Ek et al., 2006; Pronk, Palmquist, et al., 2006; Zhang et al., 2014). Pathogens and heavy metals are generally retained in the concentrate, both in case of vaporization and membrane separation (Pronk, Palmquist, et al., 2006; Liu et al., 2016). The fate of organic pollutants depends on their volatility during vaporization (Wijekoon et al., 2014), and upon membrane and pollutant properties during membrane separation (Alturki et al., 2013). Electrodialysis (ED) is another membrane separation process that has been investigated and enables the transfer of nutrients from a liquid stream (such as urine) or the liquid fraction of a slurry (such as sewage sludge) to another liquid stream (Pronk, Biebow, & Boller, 2006; Pronk et al., 2007; Tice & Kim, 2014). Desalination degrees of up to 99%

have been achieved (Pronk, Biebow, et al., 2006). Retention is high for pathogens while organic pollutants and heavy metals permeate through the membrane to some extent (Pronk, Biebow, et al., 2006; Pronk et al., 2007).

4.3.3. Phototrophic biomass growth

Through phototrophic biomass growth in aquatic or terrestrial systems, nutrients can be extracted from liquid streams, notably urine and treated effluent, and incorporated into phototrophic biomass. Algal systems have been found to have the potential to simultaneously extract N and P (as well as K and micronutrients) (Shilton et al., 2012; Vasconcelos Fernandes et al., 2015; Sukačová & Červený, 2017), but have also been found to extract organic pollutants (de Wilt et al., 2016) and heavy metals (Zeraatkar et al., 2016; Demey et al., 2018) through sorption. Moreover, algae were found to have the potential to degrade some organic pollutants (de Wilt et al., 2016; Wang et al., 2017).

4.3.4. Sorption

Sorption processes have been investigated to transfer nutrients from liquid streams (notably urine and treated effluent) to a range of carbonaceous or mineral sorbents. The sorbent itself can be the target product, or it can be an intermediary nutrient carrier from which nutrients can be transferred back to a desorption solution or regenerant. In the latter case, also synthetic sorbents/resins have been investigated.

Charcoal has been shown to have the potential to adsorb urea (Kameda et al., 2017), NH_4^+ (Cai, Qi, Liu, & He, 2016), and PO_4^{3-} (Takaya et al., 2016; Trazzi et al., 2016). Mineral sorbents generally have good cation exchange properties and good affinity for NH_4^+ and K^+ (Hedström, 2006; Jaskūnas, 2015), and have also been shown to act as precipitation nuclei for the surface precipitation of phosphates, for instance as calcium phosphate, notably if Ca^{2+} is released in exchange for NH_4^+ and K^+ (Hedström, 2006; Gustafsson et al., 2008; Karapinar, 2009; Köse & Kivanç, 2011; Guaya et al., 2016; Wan et al., 2017). Charcoal and mineral sorbents do not only remove nutrients from aqueous solutions. Charcoal, notably in the form of activated carbon, has the potential to remove some waterborne pathogens (Busscher et al., 2006), organic pollutants (Nam et al., 2014; Tong et al., 2016), and heavy metals (Kołodyńska et al., 2012) from solutions. Mineral sorbents have the potential to remove organic pollutants (Tsai et al., 2008; De Ridder et al., 2012; Chraibi et al., 2016) and heavy metals (Zorpas et al., 2000; Babel & Kurniawan, 2003; Shaheen et al., 2012; Choi & Lee, 2015) from solutions. There are indications, however, that sorbents (e.g. some types of activated carbon) can be designed to remove either nutrients or pollutants but not both.

4.3.5. Controlled precipitation

Through precipitation, crystallisation, or granulation, nutrients can be transferred from a liquid stream or the liquid fraction of a slurry to a mineral in amorphous or crystalline form. Precipitation in the Mg-Ca-NH_3-PO_4 system has been explored thoroughly (Ronteltap et al., 2007a, Ronteltap, Maurer, Hausherr, & Gujer, 2010; Marti et al., 2008; Triger et al., 2012; Liu et al., 2013; Muster et al., 2013; Vasenko & Qu, 2017). In the absence of ammonium, it is possible to precipitate the struvite analog magnesium potassium phosphate (MPP, also referred to as potassium struvite) (Wilsenach et al., 2007; Xu, Wang, Wang, & Qian, 2012; Xu et al., 2015; Nakao et al., 2017). Other studies have targeted calcium phosphate (Cunha et al., 2018), aluminum phosphate (Huang et al., 2015), ferric phosphate (Lin et al., 2017), or magnesium and sodium phosphates (Huang et al., 2015). Key mineral precipitates are described in Table S2.1 in Supporting Information 2. Pathogens may accumulate in the precipitate (Udert et al., 2006; Decrey et al., 2011; Lahr et al., 2016). Pharmaceuticals have been found to attach to the surface of precipitates rather than being incorporated in the crystal structure (Escher et al., 2006; Ronteltap et al., 2007b) and can be removed by washing (Schürmann et al., 2012).

4.3.6. Ammonia volatilisation and capture

The transfer of volatile components from a liquid to a gas stream is referred to as stripping and has long been known to be useful to remove ammonia from concentrated streams such as urine (e.g. Maurer et al., 2003; Liu et al., 2015) or ammonia-rich wastewater (e.g. Siegrist, 1996; Yuan et al., 2016). Process variations include stripping columns (e.g. Katehis et al., 1998; Antonini et al., 2011; Morales et al., 2013; Huang et al., 2015) as well as a range of setups where ammonia release is facilitated by (bio)electrochemical systems such as microbial fuel cells (e.g. Kuntke et al., 2012; Zhou et al., 2015), microbial electrolysis cells (e.g. Wu & Modin, 2013; Kuntke et al., 2014), or electrochemical cells (e.g. Desloover et al., 2012; Luther et al., 2015). Ammonia release from a liquid to a gas stream can also occur as side-effect of water extraction processes and in these cases often turns into a nitrogen loss, although the released ammonia can be captured, in principle (e.g. Horttanainen et al., 2017). Ammonia is commonly captured by wet scrubbing (i.e. absorbtion in an acid such as sulfuric acid) which renders an ammonia-rich solution (for example ammonium sulfate). Pathogens, organic pollutants, and heavy metals can be expected to largely remain in the stream from which ammonia has been released.

4.3.7. Mobilisation - separation

Prior to liquid-solid separation of slurry-like organics, release of P from the solid to the liquid fraction can be facilitated by processes such as

bioelectrochemical systems (e.g. Fischer et al., 2011; Happe et al., 2016), ozonation (e.g. Suzuki et al., 2006), additional anaerobic tanks or zones in enhanced biological phosphorus removal (EBPR) schemes (e.g. Heinzmann, 2005), or acid elution (e.g. Güney et al., 2008; Niewersch et al., 2008; Antakyali et al., 2013). Elution has also been investigated for the extraction of P from ashes. Alkaline elution dissolves phosphorus and aluminum to some extent but not iron and heavy metals; acid elution dissolves phosphorus as well as metals. Elution is often followed by processes such as membrane separation, sorption, or solvent extraction in order to separate P from heavy metals, and possibly by processes aiming at the precipitation of phosphates (Egle et al., 2015).

4.3.8. Thermal ash treatment

Separation of P and heavy metals found in ashes can be achieved through thermal ash treatment designed to release heavy metals or P from the ash, or both (sequentially) (e.g. Adam et al., 2009; Nowak et al., 2012; Schönberg et al., 2014).

4.4. Pathogen inactivation and degradation of organic pollutants

Pathogen inactivation and removal aim to ensure hygienic safety of the fertilizer product while organic pollutant degradation and removal target pharmaceuticals, pharmaceutical residues, and (synthetic) hormones. Some of the separation processes above have aimed to separate desired products (i.e. nutrients, organic matter) from those not desired (i.e. pathogens, organic pollutants, heavy metals). Treatment can also be geared towards the inactivation of pathogens or degradation of organic pollutants. Storage (e.g. Tilley et al., 2008), thermal storage (e.g. Zhou et al., 2017), and pasteurization (e.g. Lahr et al., 2016) are processes that focus on pathogen inactivation in liquid streams. These processses need concentrated streams to limit storage capacity and/or energy use. Advanced oxidation processes (Pronk et al., 2007; Lazarova & Spendlingwimmer, 2008) have the potential to achieve inactivation of pathogens (Deng & Zhao, 2015; Giannakis et al., 2017) as well as degradation of organic pollutants (Deng & Zhao, 2015). Biological treatment of liquid streams such as urine has been targeted mainly towards the degradation of organic pollutants (e.g. Abdel-Shafy & Mansour, 2016). Pathogen inactivation in slurry-like organic matter has been investigated by means of storage (e.g. Fidjeland et al., 2013), pasteurization (e.g. Forbis-Stokes et al., 2016), ammonia sanitization (e.g. Fidjeland et al., 2015), lime stabilization (e.g. Anderson et al., 2015), and desiccation (e.g. Magri et al., 2013).

5. Products

Products rendered by treatment of human excreta and streams containing human excreta can be in the form of for example energy carriers, fertilizers, or feed, while in other cases the products can be utilized for the production of biopolymers, biofuels or other high-value chemicals (Puyol et al., 2017; Chen et al., 2018). Here, we focus on products that are useful as fertilizers or for the production of synthetic fertilizers. The composition and quality of these products vary widely within the same product category. This is in part because the quality of the primary input varies between locations as a result of for example the type of industries or habits of the population, in part because different treatment trains can yield similar products (see Figure 3 and Figure S2.2 in Supporting Information 2). Below we describe different products with respect to composition and usability (Figure 5).

	NUTRIENTS					CONTAMINANTS			APPLICATION		FERTILISING TYPE					
	C	N	P	K	Micronutrients	Pathogens	Heavy Metals	Organic Pollutants[1]	Fertiliser Production	Soil Conditioner	Product Consistency: Liquid	Product Consistency: Slurry	Product Consistency: Solid	Nutrient Binding: Organic	Nutrient Binding: Mineral	Nutrient Release: Quick	Nutrient Release: Slow
Multinutrient Solution	✕	△	△	△	△	◎	◎	◉	•	•	•				•	•	
Macronutrient Solution [Urea-N]	△	△	✕	✕	✕	○	○	○	•	•	•				•	•	
Macronutrient Solution [NH4-N]	✕	△	✕	✕	✕	○	○	○	•	•	•				•	•	
Macronutrient Solution [P]	✕	✕	△	✕	✕	○	○	◉	•		•				•	•	
Macronutrient Solution [N(P)K]	✕	△	△	△	✕	○	○	◉		•	•				•	•	
Multimineral Precipitate	✕	△	△	△	△	◎	◎	◉		•		•	•		•		•
Monomineral Precipitate [Urea]	△	△	✕	✕	✕	○	○	○		•		•	•		•		•
Monomineral Precipitate [MAP]	✕	△	△	✕	✕	○	○	○	∘[4]	•		•	•		•		•
Monomineral Precipitate [MPP]	✕	✕	△	△	✕	○	○	○	∘[4]	•		•	•		•		•
Monomineral Precipitate [CaP, AlP, FeP, MgP]	✕	✕	△	✕	✕	○	○	○	∘[5]	∘[3]		•	•		•		•
Ash	✕	✕	△	△	△	○	○	◉	•	∘[4]			•		•		•
Slag	✕	✕	△	△	△	○	○	◉	•				•		•		•
Carbonaceous Sorbent (Charcoal)	⬐	△	△	?	?	◎	◎	◉		•			•		•[2]	•		•
Mineral Sorbent (Zeolite, Wollastonite, etc.)	✕	△	△	△	?	◎	◎	◉		•			•			•		•
Phototrophic Biomass (Dried Algae)	⬐	△	△	△	△	◎	◎	◉		•			•		•	•		•
Faecal-Derived Organic Matter (Sludge, etc.)	△	△	△	△	△	◎	◎	◉		•			•		•	•[1]		•

△ Recovered, losses or transfers to other products are possible.
? Potentially recovered, fate during treatment train is unclear.
⬐ Present but added from external source.
✕ Not present.
○ Not usually of concern.
◎ Possibly of concern if cotaminant reduction is insufficient.
◉ Only of concern for sewage as primary input.

1) In case of chemical P removal, P is largely bound minerally.
2) Some N as organic N in the biochar.
3) Agricultural use of CaP sometimes suggested.
4) Possibly useful.
5) AlP, FeP and MgP needs to be converted to CaP.

Figure 5. Characteristics, application potential, and fertilizing type of different product subcategories.

5.1. Nutrient solutions

Excreta-derived nutrient solutions contain nutrients derived from human excreta but are devoid of suspended organic matter. Given the wide variety of combinations of primary inputs and treatment trains that render nutrient solutions, there is considerable variation within this product category. If used as fertilizers, nutrient solutions generally are considered quick-release fertilizers as the nutrients are present as dissolved ionic species and thus directly available for plant uptake. Alternatively, some of the nutrient solutions can be used as input for the production of fertilizers. Broadly, nutrient solutions fall into two subgroups we refer to as multinutrient and macronutrient solutions. The term multinutrient solution here is used to refer to nutrient solutions that generally contain both macro- and micronutrients. The term macronutrient solution here is used to refer to nutrient solutions that contain one or several of the macronutrients NPK but no or only traces of micronutrients.

5.1.1. Multinutrient solutions

One set of recovery pathways that has received considerable attention is based on treatment trains starting from urine or yellowwater and featuring (a combination of) stabilization, contaminant reduction, and water extraction processes, or nutrient extraction processes; but the same (combinations of) processes have also been applied to other liquid streams, notably treated effluent and liquid process side streams rendered during treatment of primary inputs that contain feces. Human urine has long been known for its usefulness as fertilizer particularly rich in N (and that also contains P, K and micronutrients) (Heinonen-Tanski & Van Wijk-Sijbesma, 2005). Urine-based liquid fertilizers can be expected to be similarly useful (e.g. Bonvin et al., 2015). *Aurin* is an example of a marketable urine-based liquid fertilizer that is obtained through nitrification-distillation of hydrolyzed urine (Eawag, 2018a). Human urine has also been shown to be useful as liquid fertilizer for aquaculture (e.g. Jana et al., 2012; Rana et al., 2017), and has been investigated as input for the production of methylene urea, a slow-release synthetic nitrogen fertilizer (e.g. Ito et al., 2013). Treated effluent is useful for fertigation.

5.1.2. Macronutrient solutions (Urea-N)

Solutions rich in urea-N have been obtained, starting from unhydrolyzed urine, through sorption to and desorption from activated carbon (e.g. Ganesapillai et al., 2016; Simha et al., 2018), and through membrane separation processes, notably nanofiltration (e.g. Pronk, Palmquist, et al., 2006; Lazarova & Spendlingwimmer, 2008). In membrane separation, pathogens

and organic pollutants are retained by the membrane (Pronk, Palmquist, et al., 2006) and heavy metals are not of concern for urine as primary input. For sorption, the fate of contaminants is less reported. Also solutions rich in urea-N can be expected to be useful as fertilizer rich in nitrogen (Pronk, Palmquist, et al., 2006) and possibly as feedstock for the production of synthetic fertilizers such as methylene urea.

5.1.3. Macronutrient solutions (Ammonia-N)

Solutions rich in ammonia-N have been obtained, starting from hydrolyzed urine, through membrane separation, notably nanofiltration (e.g. Pronk, Palmquist, et al., 2006; Lazarova & Spendlingwimmer, 2008). A more widely researched approach to obtaining a solution rich in ammonia-N is the release of ammonia from liquid streams (e.g. through air stripping from urine or treated effluent) (e.g. Desloover et al., 2012; Luther et al., 2015) or organics (e.g. during thermal drying of sewage sludge) (e.g. Horttanainen et al., 2017) followed by absorption in an acid trap. Depending on the acid trap used, the respective product is ammonium sulfate ($(NH_4)_2SO_4$) (e.g. Desloover et al., 2012), ammonium borate ($(NH_4)_3BO_3$) (e.g. Kuntke et al., 2014), ammonium chloride (NH_4Cl) (e.g. Wu & Modin, 2013), ammonium nitrate (NH_4NO_3) (e.g. Horttanainen et al., 2017), or diammonium phosphate ($(NH_4)_2HPO_4$) (e.g. Licon Bernal et al., 2016). These products are generally free of pathogens, organic pollutants, and heavy metals. Yet other studies have used sorption followed by desorption to render ammonia water, starting from urine (e.g. Tarpeh et al., 2017) or treated effluent (e.g. Sancho et al., 2017; You et al., 2017). The fate of contaminants is less reported. Ammonium nitrate and ammonium sulfate are common fertilizer products applied for instance in combination with CULTAN (controlled uptake long-term ammonia nutrition) fertilization (Deppe et al., 2016). Ammonium nitrate is also a common ingredient in the production of synthetic fertilizers.

5.1.4. Macronutrient solutions (NK or NPK)

Sorption followed by desorption has also been investigated for the simultaneous recovery of NH_4^+ and K^+ from urine and their separation from Na^+ (e.g. Casadellà et al., 2016). If two or more sorbent materials are combined, sorption can also render a solution rich in in N, P and K. A less explored pathway to render a solution rich in N, P an K is HTL of wet organic matter such as feces (e.g. Lu et al., 2017). HTL transfers N, P, and K to a liquid residue while most metals (e.g. Ca, Mg, Zn, Al, Fe) are transferred to a solid residue (Lu et al., 2017). While the respective studies do not point towards a specific end use, it seems likely the macronutrient solutions would be useful as liquid fertilizers.

5.1.5. Macronutrient solutions (P)

Solutions rich in P have been obtained through a broad range of treatment trains designed to extract P from organics (e.g. sewage sludge) or inorganics (e.g. sewage sludge ash) where sewage is the primary input, as revieved extensively in Egle et al. (2015). These treatment trains generally yield a phosphoric acid, ranging from rather diluted to very pure and concentrated. Pathogens and organic pollutants are not usually of concern. Depending on the treatment train, heavy metals can be of concern, but several efforts are under way to reduce heavy metal contamination by subsequent processes such as membrane separation (e.g. Schaum et al., 2007; Parés Viader et al., 2017), sorption (e.g. Xu, He, Gu, Wang, & Shao, 2012), or solvent extraction (e.g. Hong et al., 2005). Another way to obtain a solution rich in P is through adsorption from treated effluent or other liquid process side-streams followed by desorption (e.g. Ohura et al., 2011). Phosphoric acid is rarely used for direct agricultural application but is instead commonly used in the production of synthetic fertilizers. P-rich desorption solutions seem to be most useful as starting point for the precipitation of phosphate minerals such as struvite (e.g. O'Neal & Boyer, 2013).

5.2. Precipitates

Precipitates are rendered by a wide range of processes, either as non-target (e.g. during storage) or target product (e.g. in crystallisation reactors). Common precipitates include struvite (MAP) and potassium struvite (MPP) as well as calcium, aluminum, and iron phosphates. Precipitates range from slurries consisting of individual precipitated nuclei, which can be filtered and dried to obtain a powder, to larger crystals or granules. Broadly, precipitates fall into two subgroups we refer to as multimineral and monomineral precipitates. The term multimineral precipitate here refers to precipitates that contain a range of different minerals. The term monomineral precipitate here refers to precipitates that only contain one mineral, or at least where only one mineral is the target mineral.

5.2.1. Multimineral precipitates

Treatment trains involving dehydration of urine, for instance, usually yield an inhomogeneous slurry or powder containing most of the nutrients found in the original solution, incorporated into a broad range of minerals (Antonini et al., 2012; Bethune et al., 2016; Jiang et al., 2017); where applicable also minerals originating from drying agents such as ash or lime (Dutta & Vinnerås, 2016; Senecal & Vinnerås, 2017). Multimineral precipitates have also been obtained through dehydration of anaerobic digester liquor (Ek et al., 2006). These multimineral precipitates are commonly held

to be directly useful as fertilizers (Lemming et al., 2017), although their usefulness can be hampered by high salt contents (Jiang et al., 2017).

5.2.2. Monomineral precipitates

Spontaneous precipitation of MAP or calcium phosphate (CaP) is a common phenomenon in urine collection systems (Tilley et al., 2008; Udert et al., 2003) and pipes returning anaerobic digester supernatant to the STP inflow. Treatment trains that induce precipitation through pH adjustment and/or the addition of metal ions most commonly target MAP or CaP (notably hydroxylapatite) (Melia et al., 2017), but co-precipitation of a wide variety of non-target minerals may occur (Muster et al., 2013). As amorphous precipitates may easily be overlooked, what is believed to be mostly struvite may in fact contain more other precipitates than thought, particularly in higher pH ranges (Hao et al., 2008, Hao, Wang, Loosdrecht, Van, & Hu, 2013). Other precipitates that have been targeted include MPP, AlP, FeP, and MgP. Slurries and powders tend to be less homogeneous and more prone to contain non-target minerals as well as pathogens, organic pollutants, and heavy metals. Crystals and granules can have a very high purity and homogeneity, and their quality is rather independent of the primary input and treatment train (Antonini et al., 2012); even if sewage is the primary input (and e.g. anaerobic digester supernatant the process input), heavy metal concentrations are generally lower than in commercial fertilizers (Krüger et al., 2016). The usefulness of struvite as slow-release fertilizer has long been known (Bridger et al., 1962; Johnston & Richards, 2003; Rahman et al., 2014; Talboys et al., 2016; Degryse et al., 2017). Marketed struvite pellets include Ostara Crystal Green (Ostara, 2018) and Berliner Pflanze (Berliner Wasserbetriebe, 2018). Calcium phosphate in the form of hydroxylapatite, and to a lesser extent also aluminum and ferric phosphate are held to be more useful to produce synthetic fertilizers (Melia et al., 2017).

5.3. Ashes and slags

Ashes and slags are rendered by thermal decomposition of organic matter. Ashes and slags contain nonvolatile nutrients and heavy metals incorporated in a range of minerals. These minerals are not normally further specified in the respective studies. Ashes and slags are free of pathogens and organic pollutants but heavy metals can be of concern, notably for primary inputs with higher heavy metal loads, such as sewage. Several treatment processes are under development that aim to separate P from heavy metals contained in ashes or slags, and render a decontaminated ash or slag, as described in detail in Egle et al. (2015). Ashes and slags generally are not considered a product of direct use for agriculture unless subjected to

additional treatment (Lemming et al., 2017; Melia et al., 2017). Thermochemical ash treatment, for example, has been shown to increase the bioavailability of P in the ash, making the product (calcined ash) potentially useful for direct agricultural application (Adam et al., 2008, Adam et al., 2009; Herzel et al., 2016). More commonly, however, ashes and slags are the starting point for the recovery of fertilizer products such as struvite or the production of synthetic fertilizers (Cabeza et al., 2011).

5.4. Sorbents

A wide range of sorbents has been investigated to extract one or several of the macronutrients NPK from liquid streams. Sorbents can be broadly divided into two subgroups: carbonaceous and mineral sorbents. The main carbonaceous sorbent is charcoal. Key mineral sorbents include calcinated struvite as well as aluminum silicates, calcium silicates, or calcium oxides. These sorbents are described in Table S2.2 in Supporting Information 2. Sorbents can be applied as a combined soil amendment and fertilizer (Zhang et al., 2015; Bai et al., 2017; Nakhli et al., 2017). They are generally considered slow-release fertilizers, as nutrients are released from the sorbent to soil pore water over time. Salinity potentially present in the feed solution can be reduced as sorbents have a higher affinity for desired nutrient cations (i.e. NH_4^+ and K^+) than for undesired salts cations (e.g. Na^+) (Beler-Baykal et al., 2011). Sorbents, however, are also commonly applied to remove organic micropollutants and heavy metals from aqueous solutions (Zorpas et al., 2000; Babel & Kurniawan, 2003; Kołodyńska et al., 2012; Shaheen et al., 2012; Choi & Lee, 2015). The respective bodies of literature are largely separate and studies using sorbents for selective nutrient extraction remain largely silent about potential sorption of micropollutants and heavy metals along with nutrients, as well as desorption characteristics of these contaminants. Some sorbents may also contain heavy metals to start with, for example charcoal where the feedstock is sewage sludge or sewage-derived algal biomass.

5.5. Organic solids

Organic solids include a wide variety of products that contain organic matter originating from human excreta or biomass produced during treatment of human excreta or streams containing human excreta. We here distinguish between phototrophic biomass and excreta-derived organic matter.

5.5.1. Phototrophic biomass

Phototrophic algae and cyanobacteria have received much attention in recent years and have been grown in urine and yellowater but also in liquid

streams rendered by treatment of primary inputs containing faces, such as treated effluent, anaerobic digester supernatant, or the aqueous phase after HTL. High removal of N and P from the substrate have generally been achieved (Shilton et al., 2012; Sukačová & Červený, 2017). Also heterotrophic or mixotrophic growth of microalgae has received some attention, but mostly in combination with algal biofuel production (Perez-Garcia et al., 2011). Contaminants may be of concern as algae have been shown to extract not only nutrients but also micropollutants and heavy metals from liquid streams through uptake or sorption (de Wilt et al., 2016; Zeraatkar et al., 2016; Demey et al., 2018). Algal biomass is a promising product potentially useful as plant fertilizer or animal feed (Cole et al., 2017; Wells et al., 2017). The nutrient-rich biomass is usually dried before application as a soil conditioner and fertilizer (Mulbry et al., 2005). Alternatively, it can be used as feedstock for biological decomposition (e.g. composting) or thermal decomposition (e.g. HTL).

5.5.2. Fecal-derived organic matter

Fecal-derived organic matter includes a wide variety of products that contain organic matter originating from feces or biomass produced during treatment of fecal-derived organic matter. This product type includes products that closely resemble the primary input (e.g. hygienized feces), products rendered after collection and treatment of the primary input (e.g. blackwater sludge or sewage sludge), as well as products rendered after further decomposition of aforementioned fecal-based feedstocks. These feedstocks can possibly be supplemented by other organic feedstocks (e.g. organic kitchen, yard, or wood waste) and additives (e.g. charcoal, lime, or ash) prior to (biological or thermal) decomposition. Biological decomposition renders digestate or compost (including vermicompost and fly larvae compost), whereas charcoal is the result of thermal decomposition.

These products are useful as combined soil amendments and fertilizers (Grigatti et al., 2014; Sangare et al., 2015; Kathijotes et al., 2016; Horta, 2017; Liu et al., 2018). When treatment takes place in a closed system, the product can contain N both in the form of inorganic and organic N. Treatment in open systems, however, is prone to N losses through volatilization and/or leaching. Similarly, if liquid-solid separation is applied, inorganic N can be transferred to the liquid fraction. Most N in sewage sludge and in compost in fact is organically bound and not immediately available to plants (Cogger et al., 2006; Horttanainen et al., 2017). The majority of P in fecal-derived organics is bound in mineral form. In feces, for instance, P is mainly present as calcium and iron phosphate (Rose et al., 2015). In sewage sludge, P can be present as polyphosphate incorporated in microbial biomass (biological P removal), or as aluminum or iron phosphate

(chemical P removal). P availability is variable and strongly depends on the treatment train. Sludge from biological P removal was found to be superior to precipitation with high Fe/P ratios regarding P availability and recycling (Römer, 2006; Kahiluoto et al., 2015; Lemming et al., 2017). There are indications, however, that phosphorus recovery from iron phosphate can be substantially improved by a better understanding of iron–phosphorus chemistry (Wilfert et al., 2015).

It is commonly held that satisfactory pathogen inactivation can be achieved in processes that involve exposure to elevated temperatures (Jonsson et al., 2004). Decomposition processes have the potential to fully or partially decompose organic pollutants. Heavy metals are generally of concern for (derivatives of) sewage (Tervahauta et al., 2014). As heavy metals present in human excreta were found to primarily originate from dietary sources, agricultural use of human excreta would not increase the amount of heavy metals in the food cycle (Tervahauta et al., 2014). Therefore, the importance of distinguishing black water sludge from municipal sewage sludge in sludge reuse regulations has been emphasized (Tervahauta et al., 2014). Heavy metals present in sewage sludge are mainly adsorbed to the cell surfaces of the microorganisms in the sludge (Yoshizaki & Tomida, 2000). Several studies have shown at the laboratory scale the possibility of extracting heavy metals from sewage sludge by means of acid leaching (e.g. Yoshizaki & Tomida, 2000; Naoum et al., 2001; Stylianou et al., 2007; Usharani & Vasudevan, 2016). Acid leaching, however, also dissolves phosphorus (Guilayn et al., 2017) and thus would leave a product in the form of fecal-derived organic matter depleted of heavy metals as well as phosphorus and other nutrients.

6. Patterns and trends

The synthesis presented in this review was informed by a rigorous process of organizing and and exctracting information from the pertinent literature. There is clearly no shortage of proposed recovery pathways, treatment processes, and products rendered by treatment. Here we outline a number of broader trends and patterns regarding efforts to facilitate recycling of nutrients contained in human excreta to agriculture.

6.1. Trends in process technology

For a long time, agricultural use of human excreta and streams containing human excreta was the dominant way to recycle nutrients and organic matter found in human excreta and wastewater back to agriculture (Petrik, 1954; Rockefeller, 1998; Ferguson, 2014). Beginning in the 1970s, the

extraction of nutrients started to complement recycling of the streams themselves. Early efforts include: extraction of nutrients from liquid streams through precipitation (e.g. Salutsky et al., 1972), algae growth (e.g. Mcgarry et al., 1971), or sorption (e.g. Liberti et al., 1981); and extraction of phosphorus from sewage sludge ash (e.g. Hino et al., 1998). Roughly since the mid 2000s, efforts towards nutrient extraction have intensified. Approaches that have been investigated include: extraction of nutrients from liquid streams and wet organic matter through precipitation, sorption, membrane processes, or phototrophic biomass growth; extraction of P from sewage sludge or ash; and extraction of N through various forms of ammonia release and capture (see Supporting Information 1). Also, bioelectrochemical systems have gained currency, among others to support the extraction of nutrients through electrodialysis (e.g. Zhang et al., 2013), ammonia release (e.g. Desloover et al., 2012; Wu & Modin, 2013), or precipitation (e.g. Hug & Udert, 2013). Continued research and development is taking place. For source-separated primary inputs, recent developments range from simple (e.g. struvite precipitation from urine in a simple sedimentation reactor) to more complicated approaches (e.g. bioelectrochemical systems) and include pathways that decontaminate and concentrate nutrients (to a liquid or solid product) as well as pathways based on selective nutrient extraction (notably of NPK), or a combination thereof. For sewage as primary input, recent developments are predominantly technology-intensive approaches based on selective nutrient extraction, notably of P (e.g. P leaching from sewage sludge incineration ashes). These approaches are reviewed extensively in Egle et al. (2015). The great variety of recovery pathways that involve extraction of nutrients (notably P) from sewage sludge or sewage sludge ash aligns with the trend towards incineration of a larger portion of the sludge (Kelessidis & Stasinakis, 2012; Kirchmann et al., 2017) and the anticipation of more stringent future regulation for pathogens, heavy metals, organic pollutants, and other emerging contaminants in sludge intended for land application (Mininni et al., 2015; Peccia & Westerhoff, 2015).

6.2. Focus on macronutrients NPK

The trend towards nutrient extraction coincides with a focus on the macronutrients NPK. There is no single recovery pathway that captures all nutrients and carbon in human excreta in a single product free of contamination. We see a clear divide between recovery pathways that target the recovery of (some of the) macronutrients NPK and those that more broadly target a wider selection of nutrients, and possibly also organic matter. Products containing a broader spectrum of nutrients as well as organic matter generally are less polluted, notably regarding heavy metals, when

obtained from source-separated primary inputs such as urine, feces, or blackwater. When sewage is the primary input, products containing a broader spectrum of nutrients as well as organic matter generally are prone to contain higher levels of contamination. Recovery pathways that render a product of high purity and homogeneity (e.g. macronutrient solutions, monomineral precipitates) achieve this through selective extraction of only some of the nutrients, notably macronutrients N and P. Many studies in fact do not even investigate or report the fate of K and micronutrients. For conventional urban water management and sanitation systems, and often also for new sanitation systems, the discourse generally focuses even more narrowly on P extraction and recovery. P extraction and recovery in fact is expected to become an established process in the coming decades in industrialized countries (Sartorius, von Horn, & Tettenborn, 2012).

6.3. Multiple uses for carbon

Human excreta, notably feces, contain carbon that can be valuable to improve soil quality. In conventional sewage treatment, the more readily biodegradable fraction of this carbon is usually converted into biogas and carbon dioxide through microbial metabolism. Unless sewage sludge is incinerated, the less readily biodegradable fraction of the carbon is preserved in the organic residual and potentially available to improve soil quality. But carbon may increasingly be appropriated for other purposes. Human feces and streams containing human feces can potentially serve as feedstock for the production of biocrude, bioethanol, biodiesel, biohydrogen, and syngas (Gomaa & Abed, 2017; Puyol et al., 2017; Manyuchi et al., 2018).

Likewise, feces and streams containing feces can serve as feedstock for the production of higher-value industrial chemicals, for example precursors for biopolymer synthesis and bioplastic production (Pittmann & Steinmetz, 2017; Puyol et al., 2017). These different uses of carbon do not necessarily exclude one another. But appropriation of a larger fraction of the carbon for the production of energy carriers or higher-value chemicals means that less carbon is available for the improvement of soils. The appropriation of organic matter for the production of energy or chemicals thus may to some extent compete with the recovery of organic matter to improve soil quality.

6.4. Synergies and opportunities for combining recovery pathways

While many of the studies covered in the present review target a single product with agricultural value, some studies report on a combination of recovery pathways leading to multiple products, or at least point to the possibility for a combination of recovery pathways. For example, NF of unhydrolysed urine

followed by precipitation yields a solution rich in urea and a precipitate containing N and P (e.g. Pronk, Palmquist, et al., 2006). Likewise, ammonia stripping from urine can be complemented by struvite precipitation, yielding a solution rich in ammonium and a precipitate containing N and P (e.g. Antonini et al., 2011; Wei et al., 2018). Evaporation in a vertical evaporation pipe preceded by alkalinisation yields one precipitate rich in P and a one precipitate containing the other nutrients (e.g. Eawag, 2018b). The combination of ammonia stripping, struvite precipitation, and biomass growth in a hydroponic system to treat source-separated urine even yields three products useful for agriculture: a solution rich in ammonia, a precipitate containing N and P, and a residual solution used as input to the hydroponic system (e.g. Yang et al., 2015). Other possible combinations are: pyrolysis of for instance sewage sludge and use of the char thus obtained as sorbent (e.g. Shepherd et al., 2016), or urea extraction from unhydrolysed urine through sorption in order to facilitate MPP (magnesium potassium phosphate) precipitation in the absence of N (e.g. Simha et al., 2018). Similarly, CaP granulation during anaerobic digestion of blackwater would yield three products from one reactor system: CaP, digestate, and a concentrated liquid with N, K and micronutrients.

The combination of recovery pathways can enhance overall nutrient recovery and recycling. What might be nutrient losses in a single pathway might well be captured in another product if several pathways are combined to target more than one product. This means that individual recovery pathways or products should not be judged in isolation. For example, one could argue that recovery pathways based on urine separation fail to capture about half of the P and most of the C in human excreta. Urine separation, however, does not prevent recovery of nutrients and organic matter from the stream containing the feces. On the contrary, the fact that most of the N is in the urine means that any recovery pathway starting from the stream containing the feces will be subject to lesser N losses than would be the case if urine were in this stream. Similarly, one could argue that struvite precipitation usually only captures a fraction of the P if sewage is the primary input (and anaerobic digester reject water the process input). Struvite precipitation during sewage treatment, however, does not prevent the subsequent recovery of additional P from sewage sludge or sewage sludge ashes; though systems become more complicated. Finally, individual products can also be applied in combination, for example mineral sorbents and precipitates (Lind et al., 2000; Xu et al., 2001), or compost and precipitates (Karak et al., 2015).

7. Discussion and outlook

As outlined in this review and elsewhere in the literature, a broad range of recovery technologies and pathways to facilitate recovery of nutrients and

organic matter contained in human excreta is available or under development. The two currently most mature recovery pathways are struvite crystallisation from anaerobic digester supernatant and incineration of sewage sludge with subsequent P recovery from incineration ashes. While further development and refinement of these and other recovery technologies and pathways is valuable in its own right, we believe that there is scope to ask questions that go beyond individual recovery technologies and pathways, and that better integrate end-user needs and the bigger picture.

The call for further development of technologies that recover N and K in addition to P (Mehta et al., 2015) is a step in the right direction. But in light of soil nutrient stripping (Jones et al., 2013) and soil carbon losses (Amundson et al., 2015), we think the scope of nutrient recovery should be even broader and also include micronutrients and organic matter. This will ultimately require a shift away from thinking in terms of individual recovery pathways, towards thinking in terms of sensible combinations of recovery pathways that maximize recovery of nutrients and organic matter while minimizing risks associated with contaminants.

Recognising that comprehensive nutrient recovery will again have to become a key function of human excreta management in order to help reinvigorate soil and food security, it becomes evident that a conceptual change towards framing human excreta management as part of the food cycle rather than the urban water cycle might be productive. We believe that broadening the discourse along these lines would strongly benefit from the integration of perspectives and considerations from food and farming systems with those from managing human excreta.

In other words, we argue that it is not sufficient to ask: how to recover (some of the) nutrients from (streams containing) human excreta? It is also necessary to ask: which kind of production system is envisioned as recipient of the nutrients and organic matter? How can the products best support a given production system and the achievement of specific goals such as food and soil security? Are there specific functions that need to be fulfilled by recycling from human excreta and that cannot be fulfilled by other ways of (re)cycling nutrients and organic matter? Still, there is only little research into how various recovered products fit the needs of soils and farmers (e.g. Wielemaker et al., 2018). While the present review briefly touches upon general product characteristics, further research on the effects of nutrients and contaminants from products recovered from human excreta on soil health, plant growth and human well-being would be helpful.

We recognize that recycling nutrients (and organic matter) from human excreta and streams containing human excreta to food production is only one dimension of establishing a circular nutrient metabolism where

nutrients from food are recycled back to the production of food. Establishing such a circular nutrient metabolism requires action along the entire food chain from agriculture and food processing to consumers and Waste Managementement; this includes proper management of harvest residues, animal manure, food processing residuals and waste, and human excreta (McConville et al., 2015). But nutrient recycling is currently constrained by spatial disconnects between livestock intensive areas and areas where feed is produced, and between rural areas where food is produced and urban areas where food is consumed and human excreta produced (Jones et al., 2013; Nesme et al., 2018).

Other factors that are critical for a smooth and effective transition to the widespread use of recovered fertilizer products but were not considered in the present review include legislation (Hukari et al., 2016) and social acceptance (Dahlin et al., 2016), as well as technological maturity, environmental performance, and costs (Egle et al., 2016).

We agree with Trimmer et al. (2017) that sanitation systems could become an inspirational component of societal infrastructure and an amplifiying force for sustainable development. We hope that the present review can make valuable contributions to this end, by providing inspiration to look upon the recovery of nutrients and organic matter from a broader perspective, and to better integrate perspectives from food and farming systems, the recipients of the recovered fertilizer products. The organization and classification of recovery pathways that underpins the present review could also serve as a foundation to more effectively share and consolidate what we already know about various aspects of human excreta management, and to keep track of further technological advancements.

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